Casimir forces, surface fluctuations, and thinning of superfluid films

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(Dated: February 2, 2008)

Recent experiments on the wetting of ^{4}He have shown that the film becomes thinner at the λ transition, and in the superfluid phase. The difference in thickness above and below the transition has been attributed to a Casimir interaction which is a consequence of a broken continuous symmetry in the bulk superfluid. However, the observed thinning of the film is larger than can be accounted by this Casimir force. We show that surface fluctuations give rise to an additional force, similar in form but larger in magnitude, which may explain the observations.

PACS numbers: 46.32.+x, 82.35.Rs, 87.15.La, 85.35.Kt, 07.10.Cm

Quantum fluctuations of the electromagnetic field between two conducting plates in vacuum result in long-ranged attractive interactions. This effect which has only recently experimentally verified by a series of high precision measurements[1], was first predicted by Casimir in 1948[2]. Thirty years later, Fisher and de Gennes noted that the confinement of thermal fluctuations in fluids leads to similar long-ranged forces[3]. Quite generally, geometric restrictions on a fluctuating field constrain the normal modes of fluctuations and result in fluctuation-induced or Casimir forces. These forces are controlled by correlations in the fluid; when the correlations are long-ranged, corresponding to massless fields, Casimir forces decay with distance as a simple power law[4, 5, 6, 7, 8, 9, 10, 11]. The overall strength of a Casimir interaction is typically universal. That is, it depends on symmetries of the fluctuating field, and on boundary conditions, but not on microscopic details.

An important example of a Casimir force associated with thermal fluctuations in a condensed matter system is found in ^{4}He films at and near the superfluid phase transition[12]. The finite thickness, d, of the film constrains the fluctuations of the superfluid order parameter, which then mediate a Casimir force. Experimental demonstration of this force was reported recently by Garcia and Chan[12] (GC). To produce films of various thicknesses, a stack of copper electrodes were suspended at different heights above bulk liquid helium. The thickness of the wetting layer on each electrode as a function of temperature was monitored to gauge the strength of interactions with the substrate. Figure 1 shows the change in the film thickness $\Delta d = d - d_0$, as a function of reduced temperature $t = T - T_\lambda$, near the superfluid transition point for the capacitor labeled "Cap 1" in Ref. [12]. The quantity d_0 is the thickness of the film well above the λ-point. As shown in the figure, there is a perceptible decrease in the thickness of the film at the transition point $t = 0$, followed by a substantial drop below the transition. The thinning of the film for $t \geq 0$ quantitatively confirms the theoretical predictions of attractive Casimir
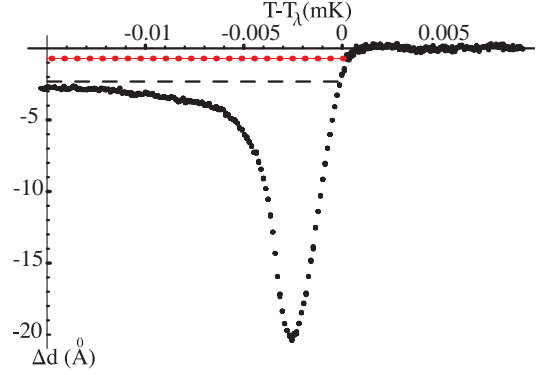

FIG. 1: The thinning of the Helium film Δd versus temperature, based on the data reported in Refs. [12, 13]. The dotted line illustrates the expected thinning of the film due to a Casimir force resulting from bulk Goldstone modes. It also coincides with the magnitude of the thinning of the film at the transition temperature as determined in Ref. [8]. The dashed line shows the expected change in thickness due to *both* bulk Goldstone modes and surface fluctuations.

interactions between parallel surfaces in the presence of Dirichelet boundary conditions[8]. At this point, no theoretical explanation has been put forth for the relatively substantial dip in the film thickness at temperatures close to and below the bulk transition temperature. In fact, the theoretical magnitude of the Casimir force at $t = 0$ is 50 times less than required to give rise to the observed maximum thinning [14].

Further below the transition point, the superfluid film partially recovers its thickness, but still remains noticeably thinner than in the normal fluid. The superfluid state, in which a continuous symmetry is broken, supports Goldstone modes that are not present in the normal phase. It is thus reasonable to expect that these long wavelength modes are responsible for the differences in thickness on the two sides of the transition point. However, the magnitude of the fluctuation-induced force associated with the Goldstone modes in the bulk of the film is too small to account for the observed reduction in

thickness in the superfluid region [14, 15] (away from criticality). The dotted line in Fig. 1 indicates the expected thinning of the film due to the Casimir force resulting from the bulk Goldstone modes.

Here, we explore the role of surface fluctuations on the thinning of a superfluid film. In particular, we investigate the fluctuation-induced force generated by the flow field in a superfluid film that accompanies undulations in the fluid-vapor interface. While it may initially appear that surface effects should be subdominant to those from the bulk, the flow fields (and hence kinetic energy) associated with surface deformations are actually quite constrained by the film thickness, d. The dependence of undulation energies on thickness leads to a contribution to the free energy that is proportional to $1/d^2$. This leads to a force favoring thinner films, with the same form as that arising from the bulk Goldstone modes, and an amplitude that is almost twice as large. The dashed line in Fig. 1 shows the expected change in the thickness of the film due to the combined influences of surface and bulk fluctuation-induced forces for a film of thickness 423 Å. As is clear from the figure, the net effect of "bulk" Goldstone modes and surface capillary modes is sufficient to explain the diminished breadth of films in the superfluid phase.

We would like to emphasize that a theoretical calculation of forces induced by surface fluctuations in liquid films is not without precedent [4, 16]. However, we are unaware of any instance in which such interactions have been detected prior to the experiments of Garcia and Chan. The key point is that in most cases, fluctuation-induced interactions (due to bulk phonons or surface modes) appear only as a small correction to the larger van der Waals forces. Traversing the superfluid transition, however, provides an instance in which the dominant atomic forces are unchanged, while the long wavelength modes due to continuous symmetry breaking can be switched on or off. Thus the *change in thickness* of the wetting film provides an ideal probe of the interactions generated by the Goldstone modes.

Deep in the superfluid state, the magnitude of the complex order parameter is fixed, but its phase ϕ can vary with relatively small energy cost. Spatial variations of ϕ are accompanied by the flow of the superfluid component. This connection underlies the two-fluid model of superfluidity[17], which leads to the mélange of acoustic and wavelike excitations supported by this system, including first, second, third and fourth sound [18, 19]. For our purposes, we decompose the variations of ϕ into components arising from bulk and surface modes. In terms of the wave-vector $\mathbf{k} \equiv (k_x, k_y, k_z = n\pi/d)$, the bulk modes have the form

$$\phi_b(\mathbf{k}) = A_{\mathbf{k}} \, e^{ik_x x + ik_y y} \cos\left(\frac{n\pi z}{d}\right). \qquad (1)$$

The cosine in Eq. (1) guarantees that $\partial\phi/\partial z$ is equal to zero at $z = 0$ and $z = d$, so that the flow field associated with phase fluctuations does not cause a displacement of either of the two interfaces. The above mode is accompanied by a superfluid velocity $\mathbf{v}_s = \hbar\nabla\phi_b/m$, and a corresponding kinetic energy $(\rho_s/2) \int d^3x v_s^2 = (\hbar^2\rho_s V/4m^2)k^2|A_{\mathbf{k}}|^2$, where ρ_s is the superfluid density, and m is the mass of a helium atom.

Integrating over all decompositions of the phase ϕ according to Eq. (1), we obtain the free energy associated with this set of bulk Goldstone modes as

$$\mathcal{F}_b = \frac{k_B T}{2} A \int \frac{d^2q}{(2\pi)^2} \sum_n \ln\left[\left(q^2 + \left(\frac{n\pi}{d}\right)^2\right)\right] + \mathcal{F}'. \quad (2)$$

Here, A is the total area of the film, and $\mathcal{F}'$ corresponds to contributions that do not generate a non-trivial dependence on d. Indeed, the sum over n can be performed by standard contour integration techniques[20]; while the dominant term is an extensive contribution to the free energy, there is an important correction that scales as $1/d^2$. The latter is the fluctuation-induced interaction, which leads to a Casimir force per unit area, of [7]

$$F_{\text{bulk}} = -\frac{k_B T}{8\pi}\frac{\zeta(3)}{d^3}. \qquad (3)$$

The thickness of the adsorbed helium film is determined by the competition of several factors, notably the loss of gravitational potential energy, and attractions to the substrate. The former can be calculated simply from the height difference h between the adsorbing plate and the bulk liquid, while the latter is due to the van der Waals interactions with the substrate[12, 21]. Fluctuation induced forces, as in Eq. (3), provide an additional component. The film thickness d is thus determined by the force balance equation

$$\frac{\gamma}{d^3}\left(1 + \frac{d}{d_{1/2}}\right)^{-1} + \frac{k_B T v}{d^3}\vartheta = mgh. \qquad (4)$$

The first term on the left hand side is the van der Waals interaction, with a leading behavior of γ/d^3 with $\gamma = 2600°$K Å^3 for a film of ^{4}He on Cu. Retardation effects due to the finite speed of light are significant for d of the order of $d_{1/2} = 193$ Å, necessitating the inclusion of the correction term. The second term on the left hand side is the Casimir force, which has the same leading behavior as the van der Waals term, with a magnitude set by $v = 45.81$ Å^3/atom and the dimensionless amplitude ϑ[22].

Unlike the van der Waals interaction, the parameter ϑ is expected to change rapidly at the superfluid transition. It is zero in the normal phase, while fluctuations of the superfluid order parameter lead to an interaction that is 'attractive' in the sense of favoring a thinner film. On approaching the transition from the normal liquid, the amplitude ϑ is a scaling function of $d (T - T_\lambda)^\nu$ (ν is the exponent for the divergence of the correlation length) which

was obtained in a two-loop Renormalization Group calculation by Krech[9]. The predicted thinning of the nearly superfluid film closely tracks the observations, e.g. as in the portion of Fig. 1 for $T \geq T_\lambda$. Currently, there are no calculations that reproduce the large dip in thickness for $T \leq T_\lambda$[24]. Well below the transition, in the superfluid phase, ϑ is expected to be a universal constant, such as in Eq. (3). The maximum amplitude calculated by Krech [8] is coincidentally rather close to the value given in Eq. (3) coming from the Goldstone modes in the bulk of the film. (Monte Carlo simulations with periodic boundary conditions in three dimensions yield a value of the critical amplitude which is roughly twice larger[26].) Thus, if these modes were the only cause for the thinning of the film in the superfluid phase, the height of the film would be roughly the same at the critical point and well below the λ-point. As indicated in Fig. 1, this is not the case. More precisely, if $h = 0.228$ cm ($mgh = 10.76$ μ°K) the equilibrium film thickness can be deduced from Eq. (4) to be $d = 423$ Å above the transition temperature. This is the thickness of the film for "Cap. 1" in the experiment of GC. In the case of the bulk Goldstone modes, $\vartheta = -\zeta(3)/8\pi$, which is too small to explain the reduction in thickness below the transition. A straightforward calculation indicates that the thinning of the film is consistent with a force which is at least three times as large as the one at the bulk critical point ($T = T_\lambda$).

To resolve the discrepancy with the experimental observation, we now resort to the effect of surface fluctuations in the superfluid[22]. According to Dzyaloshinskii, Lifshitz, and Pitaevskii (DLP), surface fluctuations in fluids also act as a source for Casimir forces[4]. However, in the case of thin liquid films, viscous damping effectively clamps the fluid, and there is no indication that such forces play any role in the thinning of the helium films.

But when the film is in the superfluid state, there is no viscosity opposing the flow fields that accompany surface deformations. To quantify the effect of surface fluctuations, we consider a film of thickness

$$d(\vec{R}, t) = d + h_{\vec{q}}(t) \, e^{i\vec{q} \cdot \vec{R}}, \qquad (5)$$

where $\vec{q} = \hat{x}q_x + \hat{y}q_y$ is the two-dimensional wave vector of the surface distortion, and $\vec{R}$ is a two-dimensional position vector. This deformation is accompanied by a distortion in the phase of the superfluid order parameter of

$$\phi_s(\vec{q}) = \frac{m}{\hbar} \, \dot{h}_{\vec{q}} \, \frac{\cosh(qz)}{q \sinh(qd)} \, e^{i\vec{q} \cdot \vec{R}} \quad . \qquad (6)$$

The form of ϕ_s is chosen such that the vertical velocity $v_z = (\hbar/m)\partial_z \phi_s$, is zero at the substrate ($z = 0$), and coincides with the motion of the liquid-vapor interface at $z = d$. Variations of ϕ_s along the z direction are exponential in qz, with $q = \pm\sqrt{q_x^2 + q_y^2}$. This choice ensures that $\nabla^2\phi_s = 0$, such that the kinetic energy is minimal,

and that there are no couplings to the bulk modes considered earlier. (We note that the velocity potential in Eq. 6 reproduces the flow field associated with the third sound mode [27].)

The kinetic energy in the flow set up by ϕ_s is

$$\frac{\rho_s}{2} \int d^3x \left(\frac{\hbar}{m} \nabla\phi_s \right)^2 = A \, \frac{\rho_s}{2} \, |\dot{h}_{\vec{q}}|^2 \, \frac{\coth(qd)}{q}. \qquad (7)$$

Surface deformations are also accompanied by a potential energy, and surface tension cost. Including these contributions, and summing over all surface modes, leads to a Hamiltonian

$$\mathcal{H}_s = \sum_{\vec{q}} \left[|\pi_{\vec{q}}|^2 \frac{q \tanh(qd)}{2\rho_s} + |h_{\vec{q}}|^2 \left(\frac{\rho f}{2} + \frac{\sigma}{2}q^2 \right) \right]. \qquad (8)$$

In the above equation, we have introduced the conjugate momentum

$$\pi_{\vec{q}} = \frac{\rho_s}{q \tanh qd} \, \dot{h}_{\vec{q}} \, ; \qquad (9)$$

in the potential energy part ρ is the mass density, σ is the surface tension, and f is the net force on the surface particles.

The classical partition function[22] associated with the surface modes is easily obtained by integrating over $\pi_{\vec{q}}$ and $h_{\vec{q}}$, leading to a contribution to the free energy of the film of

$$\mathcal{F}_s = \frac{1}{2}k_B T \sum_{\vec{q}} \left[\ln\left(\frac{q \tanh(qd)}{\rho_s} \right) + \ln(\rho f + \sigma q^2) \right]. (10)$$

Note that the contribution from the potential energy (the integral over $h_{\vec{q}}$) is present both above and below the transition, while the kinetic energy term (from integration over $\pi_{\vec{q}}$) exists only in the superfluid phase. In fact, it is only the latter that explicitly depends on the thickness of the film through the factor of $\tanh(qd)$: the d dependence of the free energy comes entirely from $(k_B T/2)\sum_{\vec{q}}\ln[\tanh(qd)]$, and is independent of various material dependent parameters appearing in Eq. (10)[28]. Taking a derivative with respect to d, we obtain the Casimir force resulting from the superfluid flow field induced by surface fluctuations, as

$$F_{\text{surface}} = -\frac{7}{4}\frac{k_B T}{8\pi}\frac{\zeta(3)}{d^3} \, , \qquad (11)$$

which is nearly twice as large as the "bulk" Goldstone mode force in Eq. (3).

The total Casimir force per unit area due to the surface fluctuations and Goldstone modes is

$$F_{\text{casimir}} = -\frac{11}{4}\frac{k_B T}{8\pi}\frac{\zeta(3)}{d^3} \approx -0.15\frac{k_B T}{d^3}. \qquad (12)$$

Using Eqs. (4) and (12), we find $\Delta d \approx 2.2$Å at temperatures slightly below T_λ, for a film whose thickness above

the transition is $d = 423$Å. The dashed line in Fig. 1 illustrates this expected change in thickness of the film, which is very close to that observed in the experiments of GC. It is important to keep in mind that the interpretation of wetting experiments on ^{4}He is complicated by the presence of microscopic scratches, dust particles and surface roughness. All these have a noticeable effect on the estimation of the thickness of the film [8, 12].

In conclusion, experiments on helium films adsorbed on copper surfaces provide a powerful tool for probing fluctuation-induced forces, as the effects due to the superfluid order parameter are turned on at the transition temperature, while the atomic forces are essentially unchanged. Fluctuations in the phase of the order parameter are an evident candidate, but can only partially account for the observed thinning of the superfluid film. We suggest that surface fluctuations provide additional interactions. In particular, in the superfluid phase the motion of the liquid-vapor interface sets up a superfluid velocity field that extends throughout the film. The corresponding fluctuation-induced force has exactly the same (material independent) form as that from the bulk phase fluctuations, but with an amplitude that is nearly twice as large. Considering the statistical and systematic errors in the experiments, it is reasonable to conclude that the combination of the bulk and surface fluctuation-induced forces is sufficient to explain the excess thinning of the film in the superfluid region. The relatively large error bars on the helium wetting experiments [12, 14, 29], do not mitigate our results, which relate to the difference in film thickness above and below the λ transition. One important extension of this work will be to find the effect of surface fluctuations on the Casimir force at—and especially immediately below—the superfluid transition.

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